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MARTIN MARIETTA

NEPTUNIUM EXPERIENCE AT PGDP

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Enrichment Technical Operations

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OPERATED BY
MARTIN MARIETTA ENERGY SYSTEMS, INC.
FOR THE UNITED STATES
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TABLE OF ACRONYMS

AEC	Atomic Energy Commission
Ci	curie, a unit of radioactivity defined as 3.7×10^{10} disintegrations per second
CIP	Cascade Improvement Program
CUP	Cascade Uprating Program
DAC	Derived Air Concentration
DOE	Department of Energy
dpm	disintegrations per minute
HPD	Health Physics Department
LLW	Low Level Waste
mCi	milli-curie, 3.7×10^{7} disintegrations per second
MTU	Metric Tons of Uranium
NLO	National Lead of Ohio
ORGDP	Oak Ridge Gaseous Diffusion Plant
ORNL	Oak Ridge National Laboratory
ORO	Oak Ridge Operations
PGDP	Paducah Gaseous Diffusion Plant
RCCP	Radioactive Contamination Control Policy
RT	Reactor Tails
RU	Recycled Uranium
TRU	Trans-Uranic material

INTRODUCTION

DOE 5400.5

A recent incident at the Paducah Gaseous Diffusion Plant (PGDP) (a spill from a drum of waste in the 746-Q building[R90-1]) has led to increased concern about transuranics (TRU) in PGDP, notably neptunium-237. Contributing to this concern is a recent rule change significantly lowering allowable ^{237}Np activity levels in the environment.[R90-3] This report is intended to provide a background summary of neptunium experience at PGDP, including historical information, operational aspects, and health physics aspects. Environmental issues are under review by a separate team, [R90-6] and will not be dealt with in this report.

^{237}Np has been of concern in the gaseous diffusion complex since the late 1950s. It is part of a larger issue relating to radionuclides in reprocessed uranium, also known as "reactor returns," "reactor tails" (RT), or "recycled uranium" (RU), which has received a good deal of attention through the years. Other radionuclides that have been of concern include other transuranics (^{239}Pu), fission products (^{99}Tc; ^{106}Ru; ^{125}Sb), other isotopes of uranium (^{232}U, ^{233}U, and ^{236}U), and daughter products of all the above radionuclides. Of the nuclides in the above group, ^{99}Tc has historically been of greatest concern in the gaseous diffusion complex. This is because the quantities fed to the cascades were large relative to other radioactive impurities and because it forms slightly volatile chemical species at cascade conditions that permit it to migrate through much of the diffusion cascade.

UF_6 feed from RU was generated on-site from UO_3 in a feed plant, and was fed during intermittent campaigns at Paducah from the early 1950s until the mid 1970s. This feed contained trace quantities of the above impurities; the presence of ^{237}Np was first recognized in this material a few years after feeding had commenced.[R56-1] Np was first detected in the isotopic cascade in 1959.[R59-2] It previously had been thought that the trace transuranics would be separated from the UF_6 in the feed manufacture process. ^{237}Np and ^{99}Tc were, for a time, recovered from feed plant waste streams for use in other Atomic Energy Commission (AEC) programs.

Reactor return uranium has not been fed since the late 1970s, although 335 MTU of unfed UF_6 from the Paducah feed plant remain on-site, and a considerable number of cylinders of commercial RU (containing about 900 MTU) have been received from Comurhex. There is continuing encouragement on the part of Department of Energy (DOE) to consider feeding reactor returns.[R89-1] The activity levels of transuranics in currently received RU are very much below standards so these materials do not appear to constitute a significant new source of Np or Pu; receipt should be re-evaluated in light of recent changes to radiological standards. Considering the many complex uncertainties, PGDP has recommended against the processing of RU unless the benefits far outweigh the costs.[R89-1]

The majority of the Np that entered the site has entered the waste streams, most of which appears to be either buried in low level waste (LLW) sites or stored in drums.

A number of comprehensive reviews of at least major aspects of the subject of reactor return transuranics have been done in the past. Recent studies include that of reference R84-1, which discusses the historical impact of reactor return feed on PGDP. Reference R86-1 is aimed primarily at discussing historical discharges, but also gives an overview of the

operations affected by transuranics and ^{99}Tc presence in the plant. Several other studies of PGDP neptunium material balance were conducted (see references R66-2; R71-1; R74-1; R76-11). These studies, and their uncertainties, will be discussed in the material balance section of this report.

Once it was recognized that neptunium was entering the Paducah cascade, studies were proposed[R59-1] and initiated[R60-1,R60-2] to study its biological effects. This apparently was the first significant biological study of the health effects of neptunium, which previously had been treated as being "similar to plutonium" based on chemical similarity and brief studies in the 1940s. Standards are the province of health physics and will be discussed in a later section. In general, specifications on TRU were established to assure that if uranium guidelines were satisfied, that TRU guidelines would automatically also be satisfied. The UF_6 feed specification on transuranics is expressed in terms of "transuranic α dis/min per gm U." Prior to 1966, this limit was 150 α dpm/gmU, which translates to 0.1 ppm Np or less (assuming no other transuranics are present). In 1966, the standard was relaxed to 1500 α dpm/gm U (i.e. 1 ppm Np or less).[R81-2]

In the various sections of this report, the quantity of Np is sometimes expressed in grams or kilograms, sometimes as disintegrations per minute (dpm), and sometimes in curies (Ci). To simplify conversion between these units, their relationship is shown in Table 1. For comparison, the properties of selected other isotopes present in the cascade are also shown. All of the isotopes shown in Table 1 are alpha emitters with the exception of ^{99}Tc, which is a beta emitter.

Table 1. Radiological properties of isotopes of interest

Isotope	Half-life	Specific Activity	
		dpm/g	~~mCi/g~~
^{237}Np	2.14×10^{6}	1.57×10^{9}	705
^{239}Pu	2.41×10^{4}	1.38×10^{11}	62,100
^{235}U	7.04×10^{8}	4.80×10^{6}	2.16
^{238}U	4.51×10^{9}	7.40×10^{5}	0.333
^{99}Tc	2.13×10^{5}	3.77×10^{10}	17,000

OPERATIONS INVOLVING TRANSURANICS

UF_6 Feed Plant

Reactor returns came primarily from the AEC facilities at Hanford and Savannah River. This material came in the form of UO_3, and was converted to UF_6 in a multi-step process in the feed plant in the C-410 building. The first steps of the process converted the UO_3 into UF_4. As this was a solid to solid process, transuranics in the original feed material would remain in the UF_4. The final step in this process was a high temperature fluorination with F_2 to produce UF_6, which was cold-trapped and transferred to feed cylinders for later introduction into the plant. Some of the UF_4 did not completely react. Solids left from this high temperature fluorination were termed "ash," and consisted of intermediate fluorides of uranium (e.g., UF_4, U_2F_9, UF_5) as well as non-volatile fluorides of impurities in the feed. Neptunium and plutonium form volatile fluorides in high concentrations of fluorine, but less readily than does uranium. A fraction of the Np originally present remained with the ash, and the remainder transferred to the UF_6 feed as NpF_6. The steel UF_6 feed cylinders would have a tendency to react with NpF_6. After feeding, the cylinder heels (i.e. residual uranium) was washed and recycled through the C-400 uranium facility. Most of the Np originally present remained in this stream. Given the poor effectiveness of uranium recovery methods for recovery of neptunium, some Np no doubt remained in the cylinders.

Only a fraction of the neptunium originally received in the UO_3, estimated to be between 10% and 40%, actually entered the cascade equipment as NpF_6. The feed plant also produced UF_6 from natural feed (i.e. "mined" as opposed to recycled uranium). The feed plant began operations in 1953 and closed in 1977, and did not operate during the 1965-1967 period.[R86-1,R88-1] Wastes from the feed plant process contained most of the Np and Pu that entered the plant. Until 1970, these waste streams were processed by aqueous chemistry methods to recover uranium. Wastes generated after 1970 at the feed plant have not been reprocessed, but have been stored.[R86-1]

Neptunium Recovery Process

The neptunium found in the RU was originally seen as a useful resource. Shortly after its discovery, a recovery facility was proposed,[R57-2] the process researched at Oak Ridge National Laboratories (ORNL),[R58-2] and a facility built at PGDP in the C-400 building.[R62-1, R62-2] The process used aqueous chemistry and ion exchange methods to recover Np from two waste streams from the feed facility (ash and cylinder washings). Production continued until about 1962; a total of about 3 kg of Np was recovered at PGDP in this campaign, and a further 1 kg was recovered at ORNL from raw material provided by PGDP. The Np recovered was shipped to Hanford; only a small quantity (9 gm) of Np remains on site at PGDP from this program.

Isotopic Cascade

A fairly small fraction of the Np received at PGDP entered the isotopic cascade. This has been estimated variously as 1 to 5 kg of Np, with analyses of materials removed from the cascade favoring lower values. It is generally assumed that, since the barrier contains well in excess of 99% of the surface area to be found within the plant, the majority of any adsorbed material will be found on the barrier. In comparative studies quantifying Np and Pu on material removed during the Cascade Improvement Program/Cascade Uprating Program (CIP/CUP), this assumption appears to be born out: typically 90% or more of the Np is found associated with the barrier.[R77-5,R77-11] One area that doesn't seem to have been considered is the feed piping. The feed system is the first cascade surfaces that UF_6 entering the plant would contact, and constitutes a potential location for deposits of reduced neptunium or plutonium fluorides.

Neptunium, as discussed in the chemistry section of this report, is relatively immobile. A survey of equipment removed from the cascade during the more recent upgrade program showed Np concentrated in the vicinity of the historical feed points for RU, several years after it had been fed to those locations in quantities sufficient to account for the material found.[R77-11] On the other hand, a small proportion of product cylinders in the late 1970s showed ppb levels of Np. Thus, there may be a very slight tendency to mobility on a time scale of decades. Most likely, the Np fed to the cascade is still in the equipment to which it was fed. Some of the converters, however, were physically relocated within the cascade, and a large number had their barrier and other cascade components removed during upgrade programs.

Ni smelting

As a result of maintenance and upgrade operations, a considerable fraction of the Np that entered the cascade has been removed. Two barrier and equipment upgrade programs took place since the 1950s. Both removed a significant fraction of the diffusion barrier (which contains the vast majority of the surface area of the cascade). The first improvement program ran from 1954 to 1961. The diffusion barrier was changed out, presumably taking a significant fraction of the Np present at that time. The barrier from the first upgrade program was shipped to Oak Ridge Gaseous Diffusion Plant (ORGDP), and, with similar material from ORGDP, was shipped to an International Nickel Company facility in the early 1960s.[R90-5]

The second program (CIP/CUP), started in 1973 and ended about 1981. Most of the barrier in the affected cascade areas was removed, taking associated deposits with it. Some equipment, however, was relocated to other areas in the plant. CIP/CUP was used as an opportunity to measure concentrations of Np in cascade equipment. Np and Pu distributions concentrated around the feed area, primarily (>70%) on barrier surfaces.[R77-4,R77-5,R77-11,R84-1]

During the CIP/CUP campaign, the barrier removed from PGDP, as well as barrier from Oak Ridge and Portsmouth, was smelted into nickel ingots[R77-1] at PGDP. These ingots were intended for sale, but failure to establish a de minimus standard for radionuclides in nickel has prevented this. The nickel ingots, as well as the slag from the process, remain on-site at

PGDP. The Np originally on the barrier separated strongly into the slag in this process.[R75-6] Furnace liners also retained elevated concentrations (on the order of 700ppb), but not large total quantities, of Np.[R82-1]

Decontamination of cascade equipment

Aqueous decontamination of cascade equipment and cylinders is used to remove uranium deposits by first dissolving and then later precipitating and filtering the solution. These decontamination processes have been designed to produce filtrates very low in radionuclides. Historically these have been discharged to the environment when below allowable standards. Sludges and filter cake historically were processed for uranium recovery at PGDP for small scale quantities, or sent to the DOE facility at Fernald for larger scale recovery. At present there are no known uranium recovery facilities operating. Filter cake currently being produced at PGDP is stored on-site.

Until about 1980, the primary decontamination process used an ammonium carbonate wash solution. This was used during the CIP/CUP program on material removed from cascade service. Laboratory tests[R77-2] showed that this method had fairly poor decontamination factors for Np (i.e. <2) compared to the factor for removal of uranium (on the order of 9). The decontamination factor is defined as ratio of original contaminant to amount remaining after decontamination. An evaluation[R76-2] of decontamination factors for barrier during CIP/CUP showed similar numbers for barrier: 9.5 for U and 1.1 for Np. Aluminum components showed similar low factors for Np, but ranged from 1.2 to 4 for decontamination of uranium.

In the 1980s, the decontamination process was changed to use sodium carbonate. Barrier and aluminum cascade components were decontaminated prior to smelting during and shortly after CIP/CUP. In these processes, decontamination factors were again on the order of 2 for Np versus factors of about 7 for U.[R83-1]

The cascade may be a continuing source for low levels of Np, primarily through decontamination operations during equipment maintenance. For example, analyses of decontamination solutions from the C-400 precipitation process shows the levels of Np during 1980 declining by 50% from its average for the period 1974 to 1980.[R81-1]

Waste Streams

Operations over the course of four decades resulted in the creation of numerous waste streams potentially containing neptunium. While it is beyond the scope of this document to attempt to identify and detail the history of all specific streams, certain categories identified as containing transuranics should be mentioned; these are summarized in Table 2.

As will be seen in the discussion of Np material balance elsewhere in this report, most of the Np that entered PGDP is to be found in these various waste streams. Surveys and inventories quantifying transuranics in these waste streams have been done in the past. PGDP transuranic material balance studies contain inventories of the locations of Np-

containing materials known on-site at the time the studies were made. One such study was done in 1971,[R71-1] and an update was done in 1974.[R74-1] The 1974 study examined, among other things, soil contamination levels in drainage ditches that had been used during early years of the plant for discharge of liquid streams potentially containing radionuclides (ditches draining C-404, C-400, and C-410 to Little Bayou). From these analyses it was estimated that less than 4 grams of Np were present in the soil of these drainage ditches.

In the UF_6 process streams, emissions are controlled by chemical trapping (passing gas streams through columns containing pellets which chemically absorb the impurities). Chemical trapping of recycled feed was generally done with MgF_2.[R75-3] This was intended primarily to remove technetium, but was also considered to be effective for Np and Pu removal. Studies have been done with CoF_2[R75-8,R82-2] for trapping of trace transuranics from UF_6 feed (in contemplation of further reactor return feeding).

Table 2. Np-containing waste and other materials categories

Feed Plant
Unused UO_3 from feed plant
"Ash" (unreacted UF_4 and intermediate uranium fluorides)
Decontamination and U recovery solutions (e.g. cylinder heels)
Feed plant hardware and material holdup therein
*UF_6 produced from feed plant but not yet fed
*Cylinders used as feed cylinders
Np recovery plant
Hardware
Waste streams (i.e. ion exchange resin; solutions; filtrates)
Decontamination Operations
Decontamination solutions
Decontamination sludges/filter cakes
Removed cascade equipment
Hardware
Barrier smelting plant
Hardware (furnace liners)
Slag
*Ni ingots

*Not necessarily to be considered as waste or surplus

Campaigns to upgrade waste handling practices have occurred several times over the years.[R72-1,R84-1,R85-1] In 1985, at the request of DOE, a study was undertaken to identify waste categories generated during the RU campaigns, and to recommend the best disposal methods. Quantities of several of the waste streams identified above are listed in Ref R85-2, and some

are not inconsequential. For example, about 1.5 tons of UO_3 were on hand as were 7.5 tons of Feed plant ash. A program had been in progress to prescribe and carry out waste treatment of TRU containing waste drums.[R85-1,R85-2] Waste treatments have been recommended for many of the stored materials at PGDP, but apparently not decided upon, so that the majority of the wastes listed in this 1985 document remain on-site in storage. Restrictive transportation requirements for TRU-containing materials have prevented transport of samples of the waste to facilities where research could be done on appropriate methods of disposal.

The recently developed "Transuranic Assessment Plan...," (attached as an Appendix), proposes to do a thorough update[R90-2] of these waste inventories as part of an effort to locate all significant TRU in PGDP. Much of the needed information appears to be available in the records of the Uranium Accountability organization at PGDP.

Reactor Return Studies

Investigations continued into the 1980s on technical problems related to continued reactor return feeding (see reference R83-2 and references therein). At that time, it was thought that reactor return feed would occur primarily at ORGDP, as (a) the material would come in the form of UF_6 (unlike the earlier Hanford and Savannah River material, which was converted on-site from UO_3), and (b) a feed trapping facility using CoF_2 had been constructed at ORGDP. Laboratory scale tests indicated a decontamination factor of 400 for NpF_6 could be achieved using CoF_2.

Based on PGDP health physics and Industrial Hygiene analysis, a total cascade content of 9 kg of ^{237}Np was, at that time, considered to be allowable at PGDP based on the then-prevailing protection standards. In the 1983 analysis, ^{237}Np in reactor return feed was considered but not regarded as a significant potential problem because (1) without trapping, many years would be required to load the cascade with its limiting quantity of Np if reactor return feed had Np at the transuranic α specification on UF_6 feed; (2) chemical trapping would be used in any case; (3) analysis of actual reactor return feed from Comurhex had a factor of 200 less transuranic α than the specification. From an impurity standpoint ^{106}Ru was considered to be more of a potential problem because no demonstrated trapping method existed at the concentrations that would be important. In any case, significant reactor return feed has not been used, largely due to concern over the levels of ^{236}U, a synthetic isotope of uranium, which of course is not amenable to chemical separation techniques from the fissionable ^{235}U. The significantly lowered environmental and health limits on neptunium relative to uranium, to levels difficult to easily and routinely detect in operation, will probably add to that concern.

The question of feeding reactor returns is by no means a dead issue. Recently, at the request of DOE/ORO, a systems analysis was conducted exploring the costs and benefits of feeding reactor returns.[R89-1] In light of uncertainty in future regulatory requirements, PGDP recommended against feeding reactor returns unless the benefits far outweigh the cost.

QUANTIFICATION OF NEPTUNIUM FLOWS AND INVENTORIES

Neptunium Flows Into the Paducah Plant Site

Neptunium-containing reactor tails material, in the form of UO_3, was received at the Paducah plant site from both Hanford and Savannah River from FY 1953 through FY 1975. However, the presence of Np in this material was apparently not recognized until 1956, with the first mention of Np (that we have been able to find) occurring in an ORNL report[R56-1] dated 3/19/56. Prior to that time, the Np content of the reactor tails is very uncertain which has led to problems (discussed below) in estimating the quantity of Np received during these early years. No reactor tails material has been fed to the Paducah cascade since September 11, 1975.

A detailed summary of all feed streams to the Paducah cascade during this time period was made in 1984.[R84-1] These data are presented in Table 3 and Fig. 1, where both the cascade feed prepared from reactor tails material and the total cascade feed are shown for each year in terms of tons of U. While the percentage of feed material made from reactor tails varied widely from year to year (as high as 65% in FY 1973), these percentage variations were largely the result of variations in the other feed materials; the feed rate of reactor tails material was actually fairly constant over most of the period, i.e., between 6,000 and 10,000 tons U per year, with smaller quantities being fed in the early years of the program and only very small amounts being fed in FY 1974 through FY 1976.

The quantity of Np received at Paducah has been estimated by several authors.[R66-1,R66-2,R71-1,R72-1,R74-1,R84-1] The estimate of 18.4 kg Np made in the latest of these documents[R84-1] is more than 4.8 kg larger than the last previous estimate of 13.6 kg.[R74-1] These figures are reconciled by the fact that the larger number includes an estimate of the quantity of material received during FY 1953 through FY 1956 for which no analytical data are available, while the smaller number neglects Np receipts during this time period. We have estimated that this accounts for nearly all of the difference in the two figures. While the true figure may lie somewhere between the two, it has been concluded that the value of 18.4 kg is as accurate a value as can be made at this time;[R90-7] accordingly, in the plant material balance discussed later we have used the figure of 18.4 kg Np, which leads to the most conservative estimates (larger amounts) of the quantity of Np unaccounted for. The estimated quantities of Np (in kilograms) received yearly at Paducah in the reactor tails material is shown in Table 3 and also in Fig. 2 (on both a yearly and cumulative basis) for FY 1953 through FY 1976.

In addition to the Np received in reactor tails material, some Np was returned from the Oak Ridge and Portsmouth sites during the CIP/CUP, associated with scrap metal (principally barrier) removed from the respective cascades and sent to Paducah for smelting and metal recovery operations. However, the quantity of Np associated with this scrap was relatively small, probably amounting to, at most, a few tenths of a kg of Np. Because of the large uncertainty associated with the quantity of Np received in the reactor tails material, as discussed above, this small additional Np input to the Paducah site has been neglected in the material balance presented below.

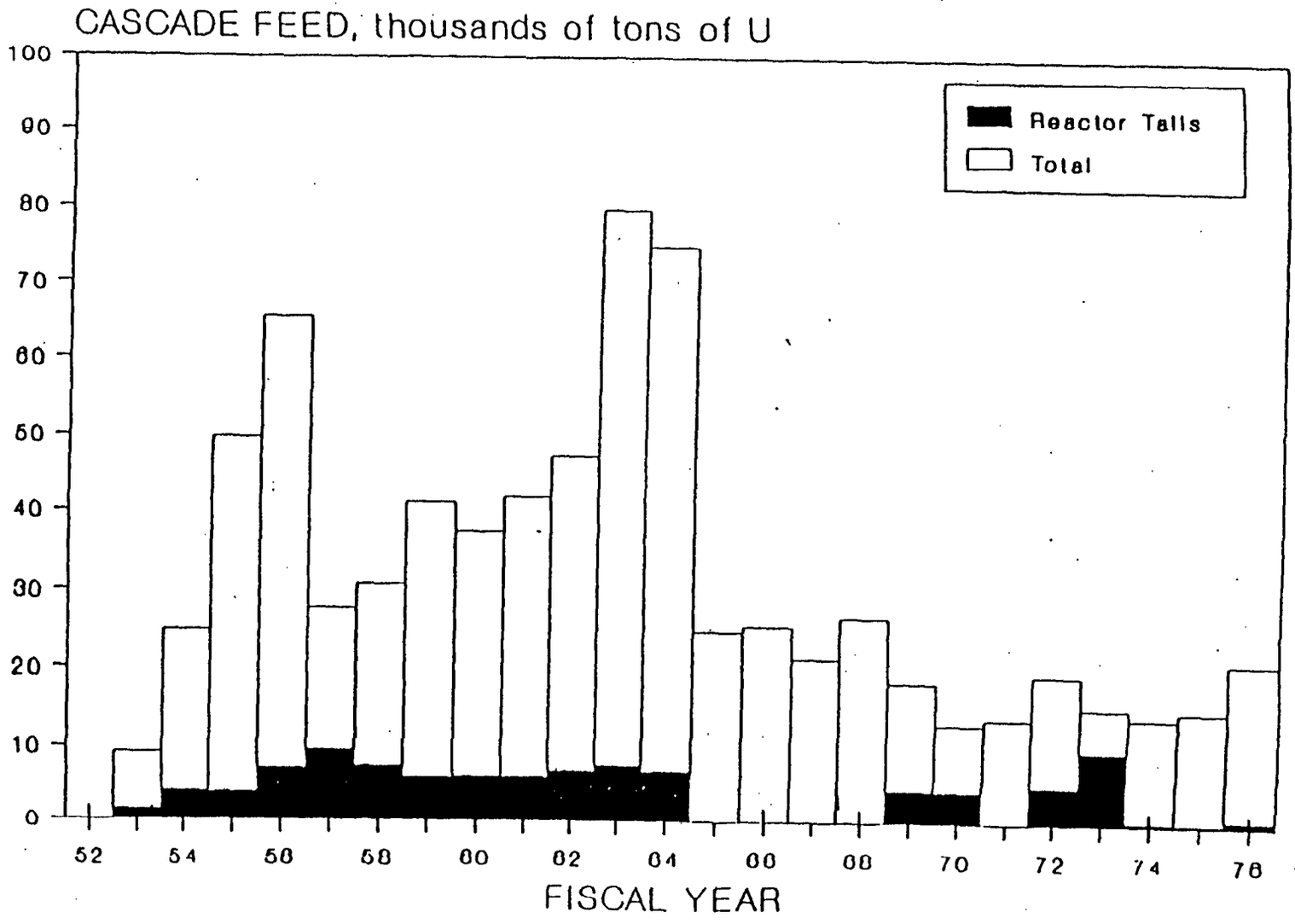

Fig. 1. Paducah cascade feed material.

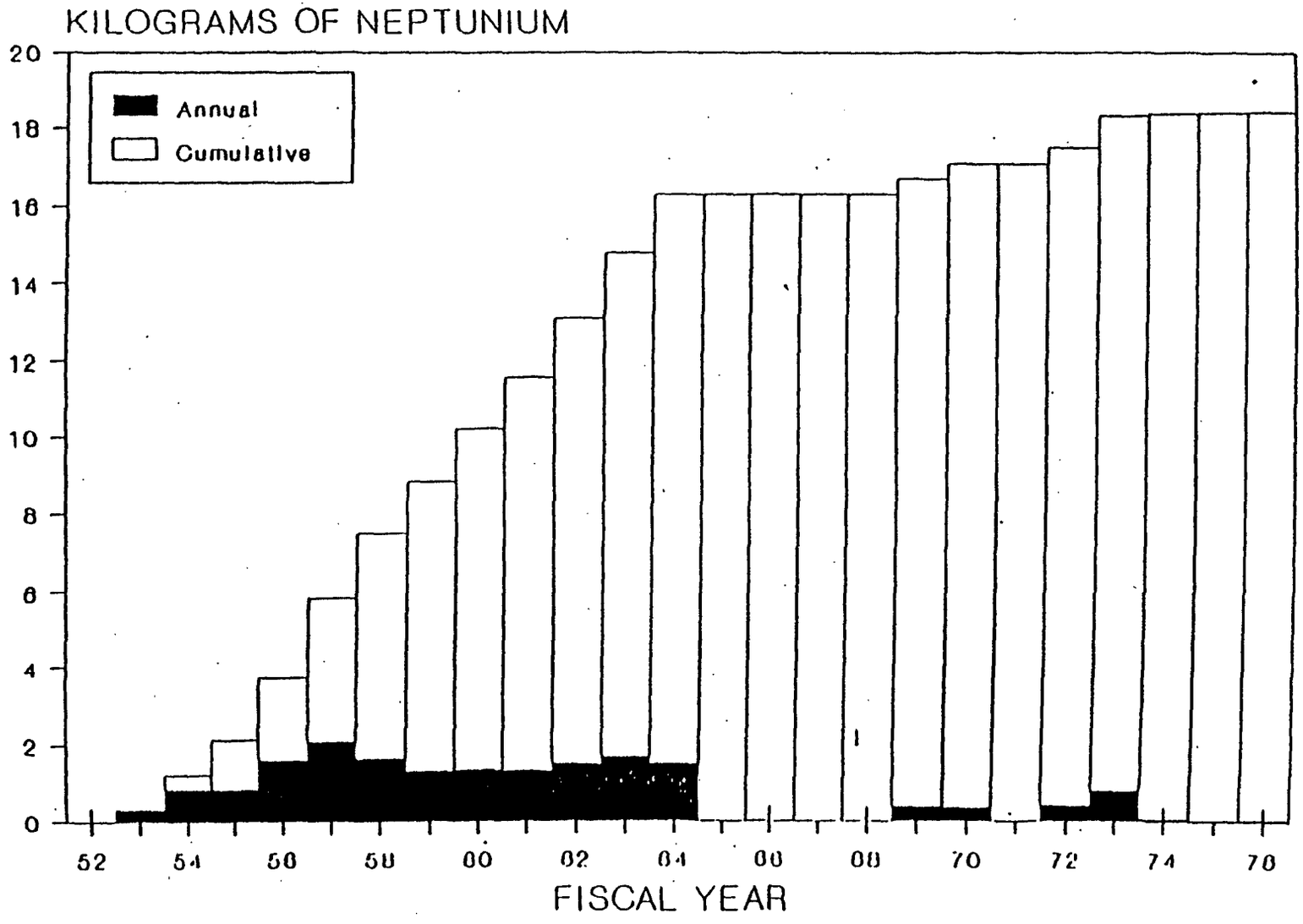

Fig. 2. Estimated neptunium received at Paducah.

Table 3. Total feed, reactor tails feed, and Np received at PGDP

FY	Total Feed tons U	Reactor Tails Feed tons U	Reactor Tails Feed % of total	Np Received kg
53	9152	1565	17.1	0.34
54	24779	4104	16.6	0.89
55	49835	4066	8.2	0.88
56	65484	7383	11.3	1.61
57	27669	9674	35.0	2.11
58	30682	7653	24.9	1.67
59	41186	6193	15.0	1.35
60	37455	6317	16.9	1.37
61	41859	6217	14.9	1.35
62	47370	6978	14.7	1.52
63	79710	7745	9.7	1.69
64	75000	7003	9.3	1.52
65	24611	0	0.0	0.00
66	25334	0	0.0	0.00
67	21251	0	0.0	0.00
68	26567	0	0.0	0.00
69	18255	4781	26.2	0.39
70	13072	4529	34.6	0.37
71	13895	0	0.0	0.00
72	19273	5283	27.4	0.43
73	15306	9904	64.7	0.81
74	14193	500	3.5	0.05
75	15023	415	2.8	0.02
76	21041	958	4.6	0.04
Total	758002	101268	13.4	18.40

Neptunium Flows Out of the Paducah Plant Site

During the period from November 1958 to October 1961[R71-1] Np was recovered from fluorination tower ash and cylinder washings to satisfy the requirements of another AEC contractor. These recovery operations were carried out partially at ORNL and partially at Paducah. A total of 4.3 kg Np was recovered and shipped from the site.[R66-1] Included in this total were 1.1 kg Np recovered from the fluorination tower ash and 3.2 kg Np recovered from the cylinder washing solutions.

Estimates have recently been published of the radionuclide releases from all of the 5 facilities operated by Martin Marietta Energy Systems (ORNL, Y-12, ORGDP, Paducah, and Portsmouth)[R88-1] and specifically from the Paducah site[R86-1] for the period 1953 thru 1987. Included in these reports are the estimated quantities of Np removed from the Paducah site in the form of liquid releases and on-site burial of solid waste (while on-site burial is not a physical flow out of the plant site, it is a well defined sink for removal of Np from the process

areas and therefore has been included in this section). These estimates are summarized, both on an annual and cumulative basis, in Figs. 3 and 4 for the liquid releases and solid burials, respectively. A total of 5.6 kg Np has been estimated to have been removed by these flow streams, 2.9 kg in the form of liquid releases and 2.7 kg as solid material buried on-site. The estimated quantity of Np released to the air was less than 0.1 kg, which is considered negligible compared to the uncertainties in the total quantities of Np received on site (see earlier discussion).

During the first cascade improvement program in the time period from 1954 through 1961, the barrier in the cascade was replaced with improved material. The barrier which was removed was ground into small flakes and returned to the International Nickel Company plant at Huntington, West Virginia.[R90-5] It is estimated below (in discussion of the plant material balance) that between 0.3 and 0.8 kg of Np was removed from the cascade in association with the barrier.

Two other paths by which small quantities of Np are known to have been removed from the plant site should be mentioned. Neptunium has been detected in a few product cylinders shipped to the other diffusion sites.[R84-1] Three cylinders were analyzed in May of 1973, and during the period FY 1976 through FY 1982 fifty-nine additional cylinders were sampled. A few of the cylinders sampled in the FY 1976A-1977 period exceeded the detectable limit of 5 ppb Np, with the highest concentration observed being 27 ppb. One of the 10 cylinders sampled during FY 1980 exceeded the lowered detectable limit of 1 ppb Np. Small concentrations of Np had earlier been detected in MgF_2 trap beds during the period of FY 1964 through FY 1966. A 10-ton UF_6 product cylinder containing 27 ppb Np (the highest observed) would contain only about 0.2 g of Np. Thus, the quantity of Np removed in product cylinders is considered to have been negligible. Measurements made on a total of 41 tails cylinders from FY 1973 through FY 1982 indicated Np levels to be below the detectable limit in every case.

Finally, some Np is known to have been contained in drums shipped to NLO (Fernald) for reprocessing of uranium.[R81-1] This material consisted of sludge from the C-400 precipitation system produced from the treatment of solutions generated during decontamination of equipment removed from the cascade during the CIP/CUP period. For the period 1/30/74 through 6/30/80, which includes a large majority of the CIP/CUP equipment decontamination, this amounted to about 0.1 kg Np. Again, this is considered a negligible quantity when compared to the large uncertainty in the total quantity of Np received at the Paducah site.

It might be noted that the flows which have been considered negligible in this and the previous section (an input of a few tenths of a kg on material returned from the other sites for smelting, and outputs of about 0.1 kg in the form of airborne releases, about 0.1 kg shipped to NLO, and an unquantified but very small quantity in product cylinders) will probably come very close to canceling each other in the overall material balance so that their omission will not effect the conclusions drawn from the results of the material balance presented in the next section.

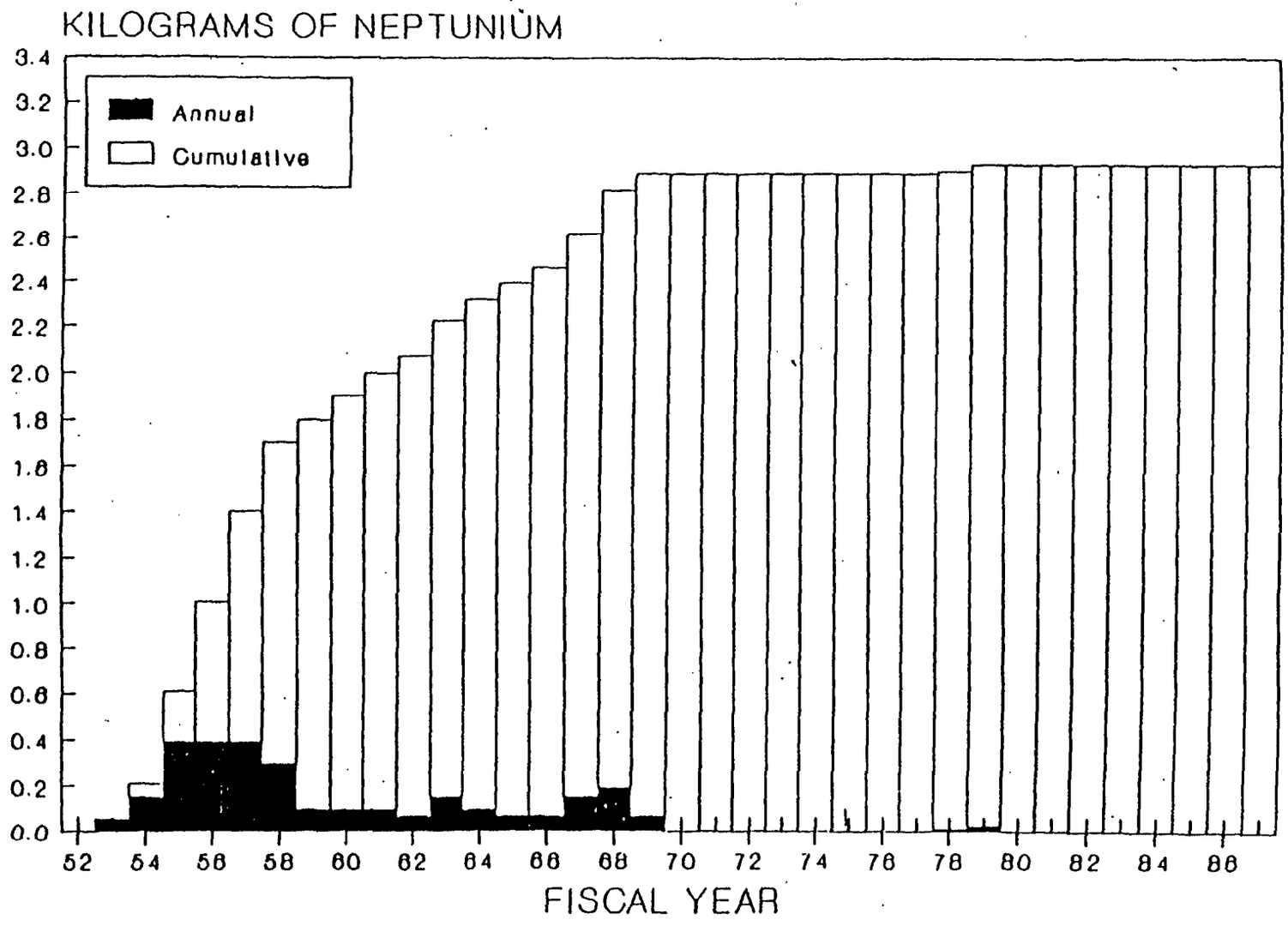

Fig. 3. Neptunium in Paducah liquid releases.

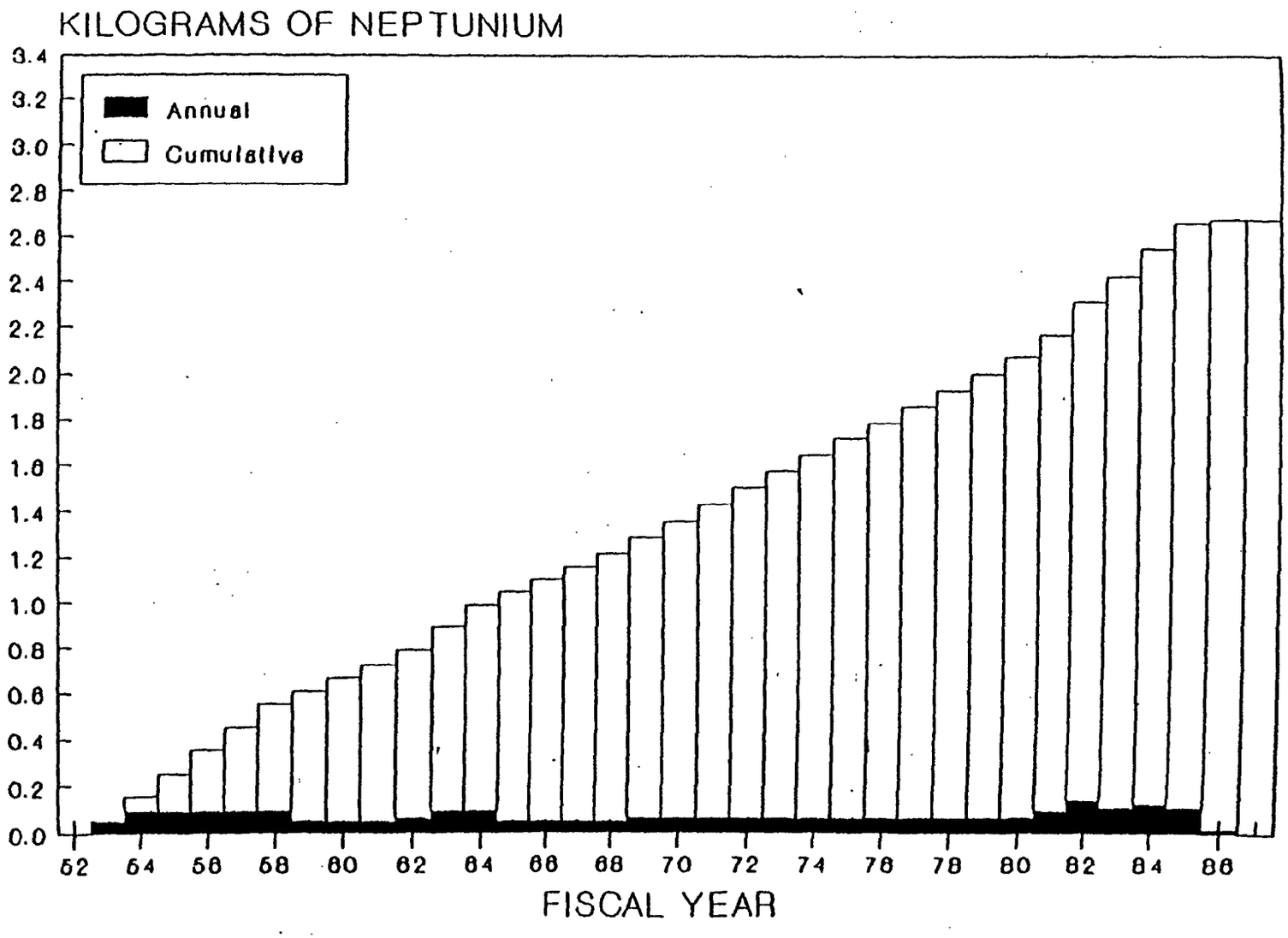

Fig. 4. Neptunium in solid waste buried on-site.

Neptunium Flows Within the Paducah Plant

A neptunium material balance for the Paducah site, including the site inputs and outputs discussed in the preceding sections as well as estimates of the intra-plant flows, is shown in Fig. 5. The methods employed to calculate the various intra-plant flows are discussed in the following paragraphs.

As discussed above, the total amount of Np received at the Paducah site has been estimated to be 18.4 kg (although the number is subject to some uncertainty) which was contained in the reactor tails material received from the Hanford and Savannah River facilities. This material was received as solid UO_3, which was then processed in the feed plant through a series of steps to convert the material to gaseous UF_6 for feed to the diffusion cascade. The first two steps involved reduction of the UO_3 to UO_2 followed by hydrofluorination to form UF_4; these were both solid-gas reactions and the Np would be expected to remain with the U through these reactions. Some dusting occurred during these reactions, and it has been estimated[R72-1] that about 5% of the Np, or 0.9 kg Np, remained in the vacuum dust removed from these systems. In the final step of the feed conversion process, the UF_4 was fluorinated to form gaseous UF_6. Unreacted UF_4, intermediate reduced uranium fluorides (such as UF_5 and U_2F_9), and a portion of the Np were removed during this process in the form of an ash residue. It has been estimated[R72-1] that 20% of the Np fed to the feed plant, or about 3.7 kg Np, was removed in this tower ash. Thus, a total of about 4.6 kg Np was removed in the process of the feed plant operations, with the remaining 13.8 kg Np being transferred along with the UF_6 into the UF_6 feed cylinders.

It is known that a substantial fraction of the Np in the product cylinders remained in the cylinder (in what was commonly referred to as the cylinder heel) after vaporization of the UF_6 into the diffusion cascade; this retained material was subsequently removed in a cylinder washing process. (Undoubtedly, some portion of the Np contained in the cylinder heels remained in the cylinders after washing; no attempt has been made in this study to quantify this remaining material or to identify the cylinders involved and their ultimate disposition.) For many years, it was assumed that 50% of the total Np received in the reactor tails material was retained in the feed cylinder (2/3 of the Np in the cylinder, since only 75% of the received Np was transferred to the cylinders), and that 25% of the received Np (1/3 of the Np in the cylinders) was fed to the diffusion cascade. This has led to an estimate of the total quantity of Np fed to the cascade of 4.6 kg,[R84-1] (25% of the 18.4 kg received).

The 25% figure seems to have originated in a 1966 document.[R66-1] A plant material balance on Np had shown that a maximum of 50% of the total Np received could have been fed to the cascade. The argument was presented that this 50% figure was too high, since analyses of dust samples from the cascade had indicated that less than 1 kg Np was contained in the cascade, which would indicate that more on the order of 10% of the Np had been vaporized into the cascade. A study had also been made in which feed cylinders had repeatedly been filled with UF_6 and then washed after a number of cycles to determine the Np present in the cylinder heels. Two series of such tests resulted in figures of 0% and 50% of the Np in the cylinder having been vaporized to the cascade. As a result of these considerations, it was decided at that time that the best estimate of the fraction of Np vaporized to the cascade was 25% (admittedly with a very large uncertainty), and that the true figure almost certainly was between 10% and 40%.

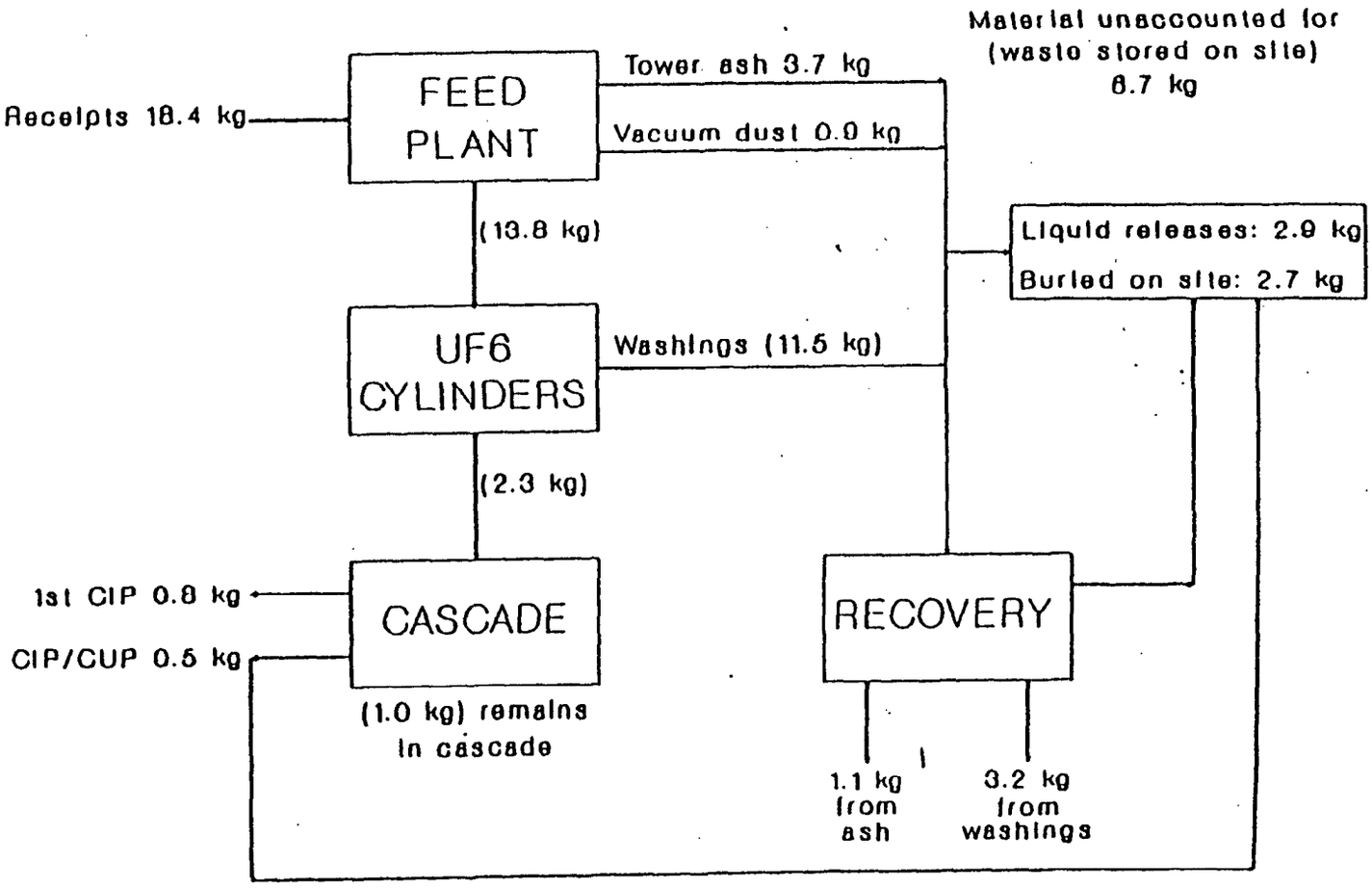

Fig. 5. Neptunium material balance.

Probably the most reliable study of this problem was published in a 1975 document.[R75-8] Based on the results of both laboratory studies and plant tests, it was concluded that at least 83% of the Np in a cylinder had been reduced and therefore not fed to the cascade. Using this figure (17% of the Np in a cylinder is fed to the cascade), the quantity of Np fed to the Paducah cascade has been recalculated; the results are shown in Fig. 6 and Table 4, which indicate that a total of 2.3 kg Np may have been fed to the diffusion cascade, although there is additional evidence (discussed below)

Table 4. PGDP cascade inventory of neptunium

FY	Np Fed kg	Total Fed to cascade kg	Removed 1st CIP kg	Removed 2nd CIP kg	Remaining after 2nd CIP, kg
53	0.04	0.04			0.04
54	0.11	0.15			0.15
55	0.11	0.26	0.03		0.23
56	0.20	0.46	0.05		0.38
57	0.27	0.73	0.09		0.56
58	0.21	0.94	0.12		0.65
59	0.17	1.11	0.15		0.67
60	0.18	1.29	0.17		0.68
61	0.17	1.46	0.20		0.65
62	0.19	1.65			0.84
63	0.21	1.86			1.05
64	0.19	2.05			1.24
65		2.05			1.24
66		2.05			1.24
67		2.05			1.24
68		2.05			1.24
69	0.05	2.10			1.29
70	0.05	2.15			1.34
71		2.15			1.34
72	0.05	2.20			1.39
73	0.10	2.30			1.49
74	0.01	2.31		0.01	1.49
75		2.31		0.03	1.46
76	0.01	2.32		0.05	1.42
77		2.32		0.09	1.33
78		2.32		0.10	1.23
79		2.32		0.12	1.11
80		2.32		0.08	1.03
81		2.32		0.01	1.02
82		2.32		1.02	
Total	2.32		0.81	0.49	

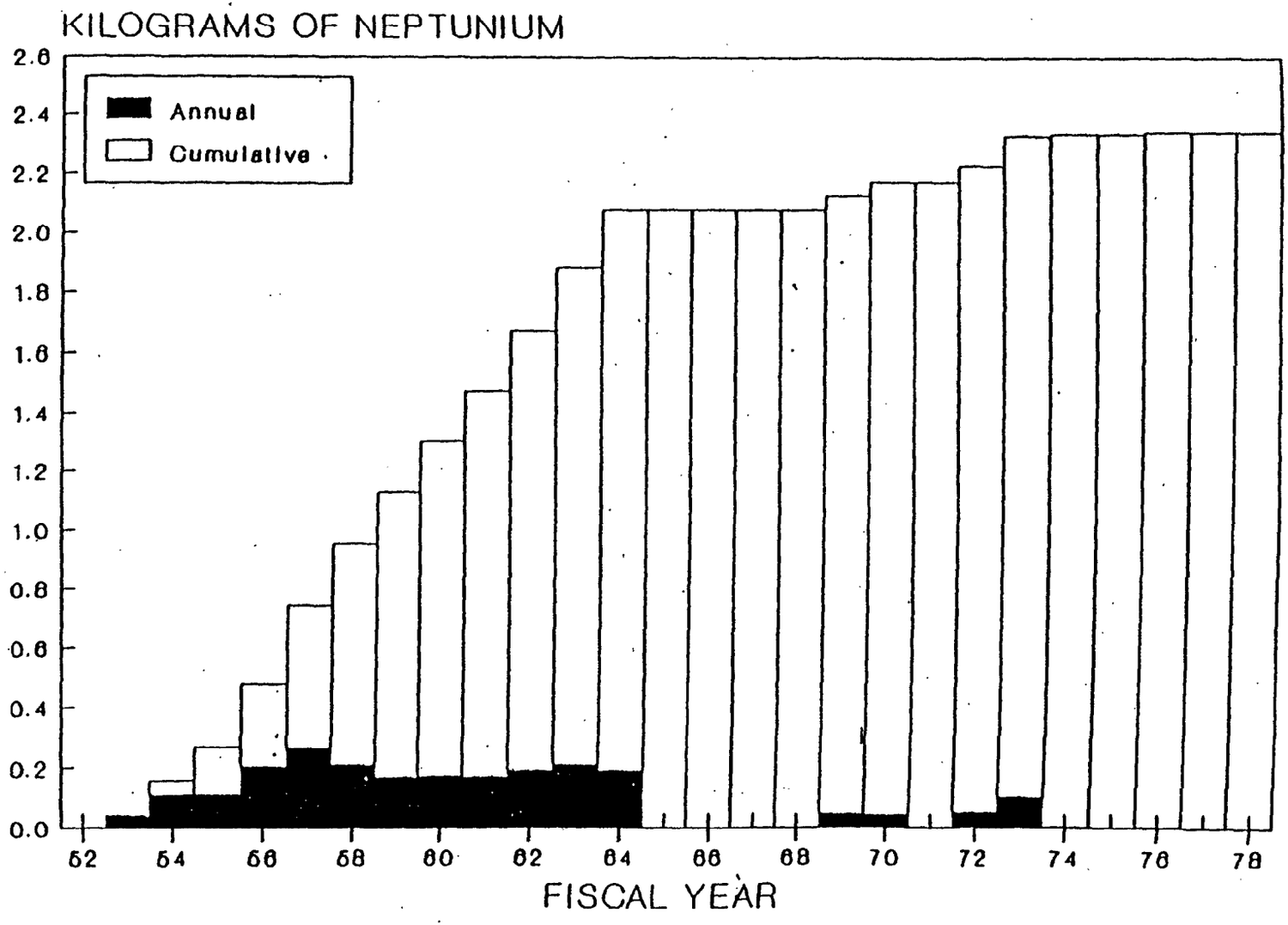

Fig. 6. Estimated neptunium fed to cascade.

which indicates that even this figure may be too large. Since no Np has been detected in the tails withdrawals and only negligible quantities have been detected in the product material from the cascade, it is concluded that essentially all of the Np fed to the cascade has been retained therein, with the exception of material removed during equipment changeouts. A portion of the Np fed to the cascade was removed during the two cascade improvement programs, during which most of the equipment in the cascade was removed and replaced with improved designs. In the first of these improvement programs, which occurred during FY 1954 through FY 1961, all of the barrier was removed from the cascade and replaced with new material. Since the barrier represents better than 99% of the total metal surface area in the cascade, it was assumed that essentially all of the Np would be associated with the barrier. To estimate the quantity of Np removed from the cascade it was assumed that 1/7 of the barrier was removed and replaced during each of the 7 fiscal-year duration of the program, so that in each of these years 1/7 of the Np present in the cascade was removed, which produced an estimate of 0.8 kg Np removed from the cascade during the first improvement program; the quantity of removed material has been included in Table 4.

The second improvement program, commonly referred to as the CIP, occurred in the period from March 1973 through September 1981. During this program, 101 of the 120 "000" cells (84%) and 52 of the 80 "00" cells (63%), were equipped with new barrier. The removed barrier was decontaminated and then sent to the Paducah metal smelter for melting and recovery of the nickel. Neptunium was recovered from the decontamination solutions using a precipitation technique. During the period from January 1974 through June 1980, which represents most of the period of the improvement program, a total of 237 g Np was recovered from the decontamination solutions.[R81-1] Assuming a decontamination factor of 2,[R76-3] this would indicate a total of 474 g Np contained on the removed barrier, so that a reasonable estimate of Np removed from the cascade during the CIP was taken as 0.5 kg; the quantity of removed material has been included in Table 4.

Removal of a total of 1.3 kg Np during the two improvement programs, coupled with the estimated 2.3 kg Np fed to the cascade, results in the estimate of 1.0 kg Np remaining in the Paducah cascade as shown in the material balance in Fig. 5. The total Np in the diffusion cascade, corrected for the material removed during the improvement programs, has been summarized in Table 4 and is shown in Fig. 7.

As mentioned earlier, there is evidence that the current cascade inventory of Np may be significantly less than the value of 1.0 kg shown on the material balance of Fig. 5. During the CIP, an attempt was made to determine the distribution of radionuclides in the Paducah cascade by the routine sampling and analysis of the equipment as it was removed from the cascade; the results of the Np analyses have been published.[R77-11] While some spreading of the Np both upstream and downstream from the feed point was evident, the results clearly show that the Np is concentrated near the feed points. From the data presented, average concentrations of 4 g Np per "000" cell and 1 g Np per "00" cell can be derived. Since 101 "000" cells and 52 "00" cells were replaced during the CIP, this would indicate that a total of 456 g Np should have been removed during the CIP, in excellent agreement with the observed total of 474 g cited earlier. These data also indicate that the cascade should currently contain only 104 g of Np. These numbers lead to estimates of 0.6 kg Np in the cascade prior to the CIP, removal of 0.3 kg Np during the first improvement program, and a total of 0.9 kg Np fed to the cascade. Thus, while the material balance of Fig. 5 indicates

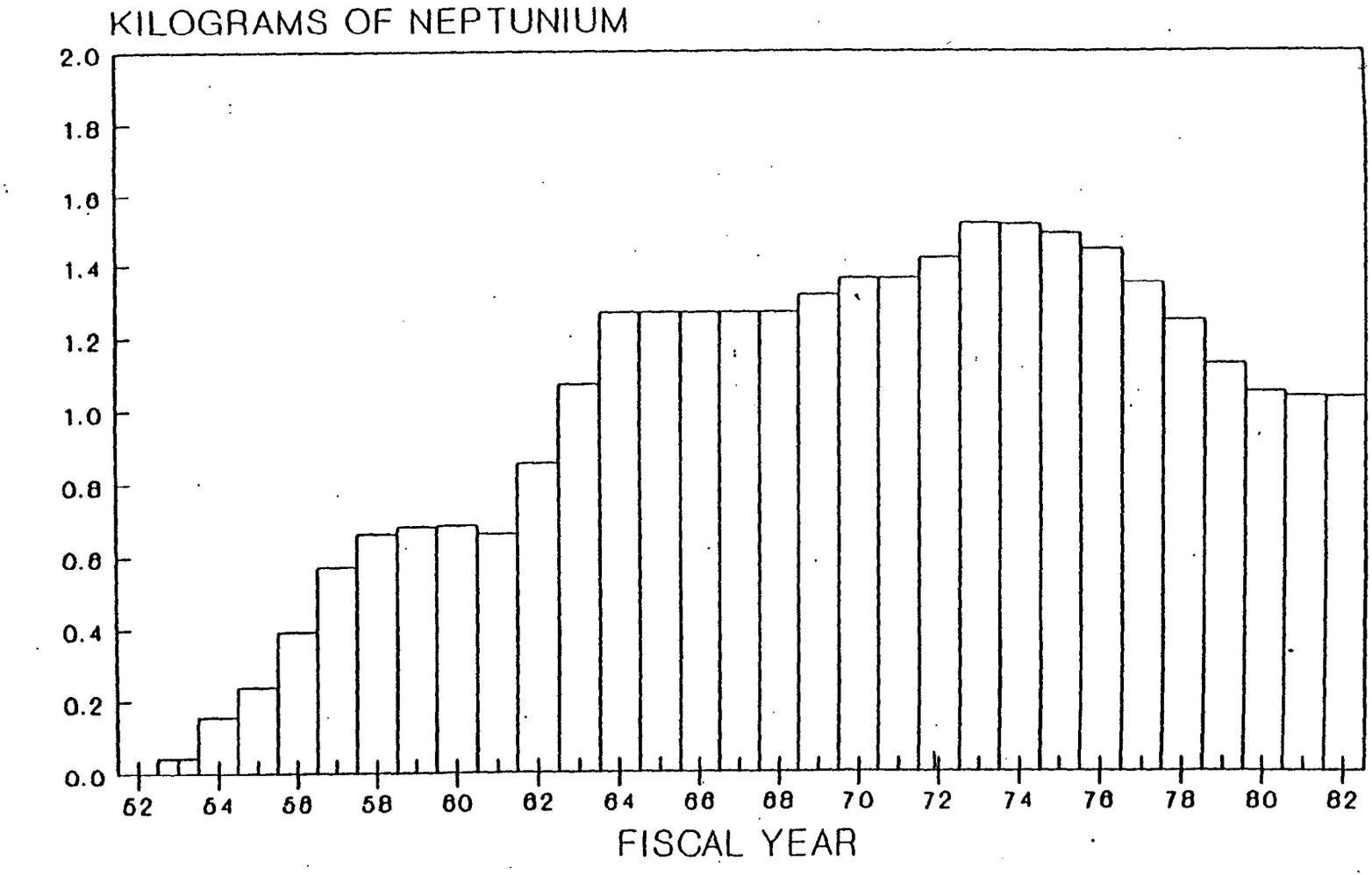

Fig. 7. Estimated neptunium in Paducah cascade.
(Cumulative)

a total feed to the cascade of 2.3 kg Np and a current cascade inventory of 1.0 kg Np, these values may be as low as 0.9 kg and 0.1 kg, respectively. A total feed of 0.9 kg Np would indicate that only 6.5% of the Np in the UF_6 feed cylinders was vaporized to the cascade, not a totally unreasonable number.

Because of the uncertainty in (1) the total quantity of Np received at the Paducah site and (2) the fraction of this total quantity which was eventually fed to the cascade, it might be well to consider several possible cases. These are summarized in Table 5.

Table 5. Results of several possible Np material balances

Case	1	2	3	4	5	6
	kg of Neptunium					
Total received on site	18.4	16.0	13.6	18.4	16.0	13.6
Removed in feed plant (25%)	4.6	4.0	3.4	4.6	4.0	3.4
Fed to UF_6 feed cylinders	13.8	12.0	10.2	13.8	12.0	10.2
Fed to cascade	2.3	2.0	1.7	0.9	0.9	0.9
Removed during 1st CIP	0.8	0.7	0.6	0.3	0.3	0.3
In cascade after 1st CIP	1.5	1.3	1.1	0.6	0.6	0.6
Removed during 2nd CIP	0.5	0.5	0.5	0.5	0.5	0.5
Current cascade inventory	1.0	0.8	0.6	0.1	0.1	0.1
Liquid releases	2.9	2.9	2.9	2.9	2.9	2.9
Buried on-site	2.7	2.7	2.7	2.7	2.7	2.7
Recovered and shipped	4.3	4.3	4.3	4.3	4.3	4.3
Np unaccounted for	6.7	4.6	2.5	7.8	5.4	3.0

In the 6 cases tabulated, three different values for the total Np received at the Paducah site are considered: (1) the maximum previously reported value of 18.4 kg[R84-1] (cases 1 and 4), (2) the minimum previously reported value of 13.6 kg[R74-1] (cases 3 and 6), and (3) the average of these two values, i.e., 16.0 kg (cases 2 and 5). For each of these three values of Np received on-site, the quantity eventually fed to the diffusion cascade has been calculated by two methods: (1) assuming 17% of the Np in the UF_6 cylinders is fed to the cascade (cases 1, 2, and 3), and (2) back-calculation of the quantity fed starting with the removal of 0.5 kg

Np during the CIP, as has been described in the previous paragraph (cases 4, 5, and 6). The results of case 1 are those which have been shown in the material balance of Fig. 5.

As can be seen from Table 5, the quantity of Np unaccounted for, which for the purposes of this document we define as material either stored in on-site storage facilities (it is known that substantial quantities of such stored material exists) and perhaps some additional losses to the environment, ranges from 2.5 kg to 7.8 kg. Perhaps the most reasonable estimate of unaccounted for material is 5.4 kg Np as shown in case 5, which assumes total receipts of 16.0 kg and a current cascade inventory of 0.1 kg.

CHEMISTRY

Reactivity in Cascade Environment

The ranking of reactivity of the volatile actinide hexafluorides is: $UF_6 < NpF_6 < PuF_6$. All three have a tendency to react with surfaces (e.g. materials of construction) to form non-volatile reduced fluorides. In common UF_6 handling practice, metals chosen for use as materials of construction are generally those which can be "passivated" by forming a stable protective fluoride layer that inhibits further reaction. The exception to this is steel, whose fluoride layer is not particularly protective but for which the reaction rate is sufficiently slow that the metal's low cost makes it attractive for moderate temperature service, most notably as UF_6 cylinders. Due to the higher reactivity of NpF_6 and PuF_6, much of the Np is left in a cylinder after feeding the UF_6, as is essentially all the Pu.

"Surfaces," as used above, can also include reduced fluorides of the more stable members of the series. In particular, NpF_6 and PuF_6 would be expected to react with UF_5 or UF_4 to form UF_6 and NpF_5 or PuF_4. Adsorption measurements conducted as part of a chemical trapping study indicated that NpF_6 did, at cascade temperatures, react with UF_4 to a degree that could not be explained by adsorption; presumably it underwent the postulated oxidation-reduction reaction. By contrast, materials likely to be found on cascade surfaces (NiF_2, CuF_2 and AlF_3) consumed NpF_6 at an area-normalized rate consistent with monolayer or partial monolayer coverage. In this study, however, difficulty was experienced with adequately passivating surfaces for use with NpF_6, and never achieved with PuF_6.[R75-8]

In addition to reaction with deposits of reduced uranium fluorides, NpF_6 is likely to react with UO_2F_2 deposits to form either an oxyfluoride or reduced fluoride of Np, liberating UF_6. As long as the cascade contains deposits of UF_5 (created by corrosion reactions of UF_6 with cascade materials of construction) and UO_2F_2 (created on reaction of UF_6 with inleaking moist air), NpF_6 should be relatively immobile. Recent campaigns to minimize such deposits may increase the mobility of NpF_6. Attempts have been made to "clean up" the cascade from the standpoint of uranium deposits. This process has included the use of off-stream treatments with fluorinating agents (F_2 and ClF_3), and the return of the reaction products of these treatments (including the UF_6 raised by the procedure as well as residual fluorinating agents. It is, therefore, possible that the mobility of NpF_6 could be increased directly (by refluorinating NpF_5 to NpF_6) or indirectly (by removal of UF_5 or UO_2F_2).

Evidence for significant mobility of Np in the cascade is equivocal but tends to support immobility. In the CIP/CUP survey (conducted on equipment as it was removed from 1975 through 1977) Np concentrations peaked in the feed area. Feeding of RU was continuing at that time, albeit at a low level, as the program proceeded, but the total quantity of Np estimated in the feed area of the cascade was much larger than the total Np fed to the cascade in the several years preceding the survey. Thus, the concentration of Np in the feed area of the cascade had to have survived for at least 5 years.

Surveys of a number of product and tails cylinders were conducted between 1973 and 1982. Of about 40 tails cylinders, none showed detectable Np (the detection limit changed from 5 to 1 ppb during this time). Of about 60 product cylinders examined, a few showed Np

above the detection limit.[R84-1] The possibility of cross contamination (e.g. reuse of a "tainted" cylinder) was not addressed. If the product UF_6 contains Np at just below the detection limit, the quantity of Np removed, if real, is minuscule (a gram or so a year). A similar rate of product stream flow (1 gm/yr) was observed in the early 60s in analysis of MgF_2 traps for Np.[R66-2] Currently, analyses for transuranics are done on one product cylinder a month. No ^{237}Np at or above the reporting limit of 5 ppb U has been detected in recent years, nor has Np been detected in recent years in chemical trap materials in the C-310 product withdrawal facility.[R90-4]

Taking these two observations at face value, one is led to the conclusion that Np in the cascade environment is very immobile, but might have sufficient mobility for a few tenths of a percent of the cascade load to leave the cascade each year through product streams.

HEALTH PHYSICS

Current Concern

Waste material containing transuranic material was released from a storage drum in the C-746-Q warehouse on March 22, 1990. The spill site was successfully decontaminated; however, the presence of TRU materials initiated an investigation into the extent of TRU materials at the PGDP facility. The investigation was designed to include an analysis of historical radiological survey data, and historical plant operations for the purpose of determining past TRU levels and likely locations of TRU material.

Personnel exposure data has historically been analyzed for uranium contamination. A re-assessment of the in vivo and in vitro data has been conducted to evaluate potential TRU exposure. Radiation workers currently involved in operations located in areas with a high potential for TRU contamination have been placed on an enhanced bioassay analysis program. Expanded in vivo and in vitro analysis has been initiated on this subset of the over radiation worker population.

Allowable limits for surface and air contamination may change substantially pending the results of a site characterization. A program has been initiated to characterize contamination in the process buildings as well as the overall site. Facility survey plans for air and surface contamination are outlined below in the section entitled "Current Actions."

Historical Studies

An evaluation of archived data by the site Health Physics Department (HPD) indicated that several evaluations of TRU materials had been conducted between the late 1950s and mid 1980s. The reports specifically discuss the influence of TRU materials on radiological work and the potential health effects associated with exposure to such material.

A Certified Health Physicist was retained by PGDP as a consultant following the March 1990 TRU contamination incident. The study was commissioned to evaluate the Health Physics program for TRU materials. Included in the project scope was a review of all available pertinent historical data, development of suggested actions necessary to assess the health impacts to employees and the public, and suggest sampling plans. The report indicated that TRU materials were identified as a potential problem as early as September 1959. Several personnel monitoring activities were initiated and concluded between the late 1950s and the mid 1980s, yet no significant exposure to personnel, based on in vivo data and pre-1988 standards were evident.

Regulatory Limits

Allowable limits for contamination and exposure are defined in more detail today than at any time in the history of radiation protection. The issuance of DOE Order 5480.11 (order) on 12-21-88 and the DOE Oak Ridge Operations Radioactive Contamination Control

Policy (RCCP); Revised 10-89 have provided guidance for personnel exposure and facility contamination which has a large impact on the conduct of operations at DOE facilities.

The PGDP facility has been operated as a "Uranium" facility since the issuance of the order. Confirmed presence of TRU material following data review of the current assessment program will result in operational changes to release limits for personnel and property, as well as Derived Air Concentrations (DAC). The size of airborne radioactive areas and subsequent respirator usage will be increased due to a 1,000 fold decrease in the allowable DAC for TRU materials versus the DAC for uranium. The size of contamination areas will increase due to a factor of 50 decrease in surface contamination limits.

Current Actions

A survey plan has been developed to evaluate the presence and extent of TRU contamination at PGDP. The plan (attached) "Transuranic Assessment Plan for Paducah Gaseous Diffusion Plant" encompasses a review of historical data, as well as a three step survey plan based upon potential contamination. The survey scope includes facility air and surface contamination, and an expanded personnel dosimetry analysis. An implementation schedule is included for phase one of the assessment plan.

Additional short term support has been procured in order to completely evaluate the TRU concerns at PGDP. Analytical laboratory support for analysis of air and surface contamination, and bioassay samples is provided via a sub-contract. Health Physics technical resources have been coordinated through two consulting organizations. The quantity of instruments available for facility air and surface contamination characterizations has been enhanced and personnel egress monitoring equipment has been supplied by other Energy Systems facilities.

The release of material in C-746-Q on March 22 1990 occurred as a result of improper transportation technique and inadequate facility design. Modifications to the drum movement procedures and facility upgrades have been recommended.

Continuous job coverage by the Health Physics Department (HPD) has been instituted to assist in the future characterization of TRU concerns and personnel protection for jobs which involve:

- UF_6 process system breaks
- Seal changes/rocker assemblies and motor coupling removal
- Welding, grinding, or buffing on UF_6 process related equipment

jobs with the potential for high air borne concentrations.

Related Regulatory or Health Concerns

Specifications on quantities of transuranics in reactor return uranium in the past were devised to assure that radiological limits and handling practices for uranium would automatically satisfy similar standards for transuranics. For example, water release standards

for neptunium and plutonium until very recently were higher in terms of activity (i.e. disintegrations per unit time per unit volume) than for uranium (per DOE Order 5480.1A, ^{237}Np was $3x10^{-6}$ mCi/l versus $6x10^{-7}$ mCi/l for ^{238}U). A very recent change, DOE Order 5400.5,[R90-3] lowered the allowable discharge level for ^{237}Np to $3x10^{-8}$ mCi/l while leaving ^{238}U the same. Thus, while it formerly was valid to control to the uranium activity levels, the rule change (which was to take effect in May of 1990) makes this no longer true. Similar problems arise with natural daughter products of uranium.

A number of issues have been identified during the recent attention given to Np in the PGDP. Many of these are covered in the action plan for radiological assessment. These deal largely with contamination control and health physics controls. One that is not is the subject of heat stress. Since early May, probably prompted by new limits for airborne contamination of neptunium and certain daughter products of natural uranium,[R90-2] maintenance activities on open equipment in the PGDP cascade have been done with "head-to-toe" protective clothing (formerly, only respirators were required). The cascade buildings typically are in excess of 100° F as it is; fully suited workers in this environment face the potential of heat injury. Typically, workers have been able to work about 15 min in this environment before taking 45 min off to recover from the high temperatures. Industrial hygiene and medical department personnel are giving this matter their attention; the possibility of air conditioned suits is being considered. In the present situation, however, it is not clear that the overall safety of the employees has been improved by these protective measures.

CONCLUSIONS

Conclusions that can be drawn from this study are as follows. The presence of transuranic contamination and the associated health physics implications have been recognized at the Paducah plant since the 1950s. While procedures were instituted that led to effective simultaneous control of uranium and TRU contamination, recent DOE order changes may require a significant revision to past contamination control practices, and the presence of transuranics requires significantly different control procedures.

Much of the information described in this report as "not determined" is probably not "lost to history," and a more thorough review of the available historical material may reveal more information about these subjects. A more complete data review appears to be a part of the overall action plan (reference R90-2). This should assist in prioritization of disposal or consolidation efforts.

A few potential locations where transuranic residues may occur have been discussed in this report that were not explicitly mentioned in earlier studies. These include residues in cylinders historically used for containing feed produced from RU, and the cascade feed facility's associated plumbing.

It should be noted that ORGDP also had a feed plant which produced UF_6 from RU, although in quantities significantly smaller than at PGDP. Similar (but probably smaller-scale) TRU concerns may, therefore, apply to ORGDP as well as at PGDP.

Though this report has dealt primarily with neptunium experience at PGDP, the entire range of radiological hazards should be considered as an integrated and balanced whole. These hazards include TRU, U-isotopes (including ^{232}U, ^{233}U, and ^{236}U), fission products, and the daughter products of the above. Under the new guidelines, certain daughter products, such as ^{230}Th and ^{231}Pa, may be of as much concern as TRU materials.

Finally, in defining the protective measures necessary, care must be taken to assure that those measures do not themselves jeopardize the health and safety of employees.

ACKNOWLEDGEMENTS

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APPENDIX

The following is the text of the "TRANSURANIC ASSESSMENT PLAN for the Paducah Gaseous Diffusion Plant," which was developed in response to the recent TRU concerns.

TRANSURANIC ASSESSMENT PLAN FOR PADUCAH GASEOUS DIFFUSION PLANT

I. HISTORY

Large quantities of recycled uranium (reactor returns) from Department of Energy (DOE) programs at Hanford, Washington, and Savannah River, South Carolina were introduced into the process feed system at the PGDP from its startup in 1952 until the mid 1970s. These reactor returns contained transuranic (TRU) elements which were formed during the irradiation of the original fuel elements. The most important TRU materials from a personnel exposure perspective are ^{237}Np and $^{239/240}Pu$.

Most of the contaminants were removed during chemical reprocessing, but plutonium and neptunium carried through the uranium recovery process and were introduced into the cascades during the UF_6 feed process. The amount of TRU materials in the feed cylinders was characterized, but recent sampling indicates that TRU contaminants introduced into the process lines may be higher than previously estimated. In the mid 1970s a major effort was initiated to upgrade the PGDP cascade facilities. Improvements included replacement of most of the gaseous diffusion barrier. This occurred during the time the last TRU material was fed and after the last recycle of uranium had been fed through the plant. Removal of the barrier was assumed to have reduced the TRU inventory in the process system, but there is no data which indicates that the surveys were compared to TRU release limits.

II. PURPOSE

This survey plan is designed to assess TRU materials and the associated radiological hazard at the Paducah Gaseous Diffusion Plant (PGDP).

Sampling has been conducted on various process equipment, process materials, airborne and waterborne radiological emissions, and the workplace during the 1970s and 1980s. These surveys did not address the presence of TRU materials by using appropriate survey instrumentation procedures nor release guidelines. This survey plan is designed to provide information on the presence and quantity of TRU materials at the PGDP.

III. SURVEY SCOPE

This Transuranic Assessment Plan will be conducted in phases, with the scope of each phase determined by the results of the previous survey activity. This plan will discuss the first phase of the survey activity in detail with later phases only generically described.

A. PHASE 1 SURVEY PLAN

The purpose of the Phase 1 survey will be to review historical data, establish sampling criteria for process radioactive materials, identify workplace areas with potential TRU concerns and provide radiological characterization of TRU levels in those areas, evaluate specific health physics requirements for personnel protection, and establish monitoring requirements for workplace and personnel.

1. Workplace Evaluation and Sampling

Samples of uranium materials and process solutions will be collected from the workplace to determine the presence and ratio of TRU activity to uranium activity. The sampling of the workplace environment is prioritized based on the potential radiological hazard from TRU materials, based on number of personnel in each facility and operational activities.

Group I		Schedule
• C-410/420	Feed Plant & Expansion	04/23/90*
• C-400	Cleaning Building	05/09/90*
• C-720	Maintenance & Stores Building	05/16/90*
• C-746	Warehouses	06/08/90
• C-310	All Floor Cells 2,4, & 6	05/15/90*
• C-333	Process Building	06/08/90
• C-337	Process Building	04/17/90*
• C-335	Process Building	06/08/90
• C-331	Process Building	06/08/90
• C-409	Stabilization Building	04/19/90*

* Survey and laboratory analysis completed.

Group II		Schedule
• C-333-A	Feed Vaporization Facility	06/15/90
• C-337-A	Feed Vaporization Facility	06/06/90*
• C-710	Technical Services Building	05/30/90*
• C-310	Product Building (Remaining)	06/08/90
• C-315	Surge & Waste Building	05/30/90*
• C-620	Compression Building	05/30/90*
• C-750	Garage	05/30/90*
• C-360	Toll Transfer & Sampling Building	05/30/90*
• C-200	Guard & Fire Department	05/30/90*
• C-102	Medical Facility	06/15/90

* Survey and laboratory analysis completed.

Group III		Schedule
• C-300	Central Control	06/22/90
• C-302	Operations Administration Building	06/22/90
• C-724	Carpenter Shop	06/22/90
• C-340	Conversion Facility (Shut Down)	05/30/90*

* Survey and laboratory analysis completed.

- Samples of spray booth wash solutions will be collected to provide an estimate of the material present in cascade equipment.

- Process gas (PG) inventory samples will be collected to provide information regarding the potential for TRU contamination of the workplace through PG releases.

- Analyses of process vent samples will be performed to identify any detectable TRU in emissions to the environment.

- Wipe samples of internal cascade equipment surfaces will be collected to provide additional information regarding TRU contamination levels in various cascade locations, concentrating on C-333 and C-337.

- Selected samples collected at the PGDP site will be sent to an independent laboratory for confirmatory analyses.

- Samples will be collected from decontamination buildings and uranium recovery areas, specifically:

  Cylinder wash solutions
  Raffinate from uranium recovery

2. Evaluation of Workplace Radiological Survey Data

   a. Workplace Contamination Surveys

The process data on TRU contaminant levels will be evaluated to determine the presence of TRU material. Based on this data, an evaluation will be conducted to determine whether the current radiological controls are adequate for the level of TRU contamination. Current workplace monitoring data will be evaluated to determine whether additional sampling specific to TRU contamination will be required. The TRU contamination found in the process materials will be used to determine any changes to the survey and posting requirements for radiologically controlled areas. Implementation of modified facility controls and survey methods will occur on a phased schedule based upon contamination levels, occupancy, and facility use.

b. Workplace Air Contamination Monitoring

The PGDP facility has ~25 continuous passive air monitors located in various areas of the workplace. These samples are changed daily and counted for gross alpha and gross beta activities. Air filters which have 4.4 dpm/m^3 or more of alpha activity will be analyzed for TRU materials. This action level was chosen based on the DAC for neptunium which is 4.4 dpm/m^3.

c. Personnel Protection

Personnel protection requirements will be evaluated based on the ratios of TRU to U activity found in each area/process. Current requirements have been specified based on health physics evaluations of the work activity, representative air sampling, and surface contamination monitoring. In general, PGDP controls are based on 10% of the DAC for the most restrictive radionuclides present on the sample, and the contamination levels specified in DOE Order 5480.11, Attachment 2.

Selected personnel will be sent to the Feed Materials Production Center, Fernald, Ohio, for confirmatory in vivo analysis. Additional personnel may be added to the program pending the results of the initial study.

d. Dose Assessment

The PGDP site currently performs in-vitro and in-vivo analyses for exposed and potentially exposed personnel. The in-vitro analysis includes total uranium and technetium analyses; the in-vivo count includes ^{235}U, ^{238}U, ^{237}Np, ^{99}Tc, and other radionuclides. Whole body counting can determine long lived deposits of ^{237}Np, but should not be used for current dose control of employees. Urinalysis is a better method of detecting low levels of ^{237}Np due to its excretion rate, complemented by WBC data. The criteria in the DRAFT DOE bioassay standard will be used to perform this assessment. Urine samples will be analyzed by sub-contract laboratory.

3. Schedule

• Initiate survey plan	05/21/90
• Complete survey	06/22/90
• Complete sample analyses	08/17/90
• Submit draft report to Program Manager, ES Health Physics	08/31/90
• Submit final report to Program Manager, ES Health Physics	09/28/90

B. PHASE 2 SURVEY PLAN

The purpose of the Phase 2 survey is to further characterize those areas identified in Phase 1 which have known TRU contamination. A detailed sampling plan will be developed to fully assess each building.

C. PHASE 3 SURVEY PLAN

The purpose of the Phase 3 survey is to characterize those areas of the PGDP which were not characterized during the Phase 1 effort. These facilities will be characterized in accordance with the site implementation plan for DOE Order 5480.11.

DISTRIBUTION

Oak Ridge Gaseous Diffusion Plant

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